toxics

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Supplementary Materials: Necessity of Assessing Biological Exposure to Arsenic Species by Two Representative Analytical Methods

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Table S1. Adjusted geometric mean concentrations of As species in 102 abandoned metal mines.

	n (%)	Adjusted GM (95% CI) (μg/g cr)							
		Inorganic As			Organic As			AsIII + AsV + MMA + DMA	HG-AAS
		AsIII	AsV	Subtotal	MMA	DMA	Subtotal		
Total	442 (100.0)	0.10 (0.08–0.13)	0.31 (0.24–0.39)	0.57 (0.45–0.72)	0.90 (0.65–1.23)	68.75 (61.61–76.73)	72.19 (64.85–80.37)	74.85 (67.50–82.99)	55.07 (48.41–62.64)
Sex									
Male	182 (41.2)	0.13 (0.10–0.17) [a]	0.35 (0.26–0.47) [a]	0.72 (0.55–0.94) [a]	1.09 (0.76–1.58)	59.05 (51.95–67.13) [a]	62.50 (55.14–70.84) [a]	65.98 (58.48–74.43) [a]	44.74 (38.49–52.01) [a]
Female	260 (58.8)	0.08 (0.05–0.11) [b]	0.27 (0.18–0.39) [b]	0.46 (0.32–0.64) [b]	0.74 (0.46–1.18)	80.05 (68.02–94.21) [b]	83.40 (71.12–97.79) [b]	84.91 (72.85–98.98) [b]	67.77 (55.98–82.06) [b]
p-value		0.008	0.256	0.030	0.168	0.002	0.003	0.007	<0.001
Age (y)									
≤59	77 (17.4)	0.16 (0.11–0.24) [a]	0.48 (0.31–0.74) [a]	1.06 (0.72–1.58) [a]	0.80 (0.47–1.37)	49.77 (41.31–59.96) [b]	52.68 (43.91–63.20) [b]	56.48 (47.40–67.31) [b]	39.62 (31.83–49.31) [b]
60–69	140 (31.7)	0.10 (0.08–0.14) [a]	0.36 (0.26–0.50) [a]	0.65 (0.48–0.88) [ab]	0.98 (0.65–1.48)	62.46 (54.08–72.14) [ab]	65.96 (57.30–75.94) [ab]	68.89 (60.15–78.90) [ab]	47.48 (40.08–56.23) [ab]
70–79	154 (34.8)	0.08 (0.06–0.11) [a]	0.23 (0.16–0.32) [a]	0.39 (0.28–0.53) [b]	0.82 (0.54–1.25)	76.59 (66.10–88.75) [a]	80.02 (69.29–92.42) [a]	82.12 (71.49–94.33) [a]	60.84 (51.17–72.33) [a]
≥80	71 (16.1)	0.08 (0.05–0.12) [a]	0.22 (0.14–0.35) [a]	0.40 (0.26–0.61) [b]	1.01 (0.56–1.81)	93.84 (76.56–115.04) [a]	97.69 (80.06–119.20) [a]	98.23 (81.10–118.98) [a]	80.36 (63.27–102.07) [a]
p-value		0.018	0.013	<0.001	0.824	<0.001	<0.001	<0.001	<0.001
Period of residence (y)									
≤20	85 (19.2)	0.09 (0.06–0.13) [ab]	0.21 (0.14–0.32)	0.38 (0.26–0.55) [b]	0.55 (0.33–0.92) [b]	69.73 (58.31–83.39)	72.65 (61.00–86.53)	73.95 (62.50–87.51)	61.54 (49.88–75.93) [a]
21–40	69 (15.6)	0.07 (0.05–0.11) [b]	0.32 (0.20–0.51)	0.49 (0.32–0.74) [ab]	0.70 (0.39–1.25) [ab]	74.46 (60.89–91.06)	77.65 (63.79–94.53)	79.83 (66.06–96.48)	59.22 (46.76–75.01) [a]

41–60	152 (34.4)	0.14 (0.11–0.20) [a]	0.30 (0.21–0.42)	0.72 (0.53–0.98) [a]	1.41 (0.93–2.14) [a]	72.69 (62.90–84.00)	76.94 (66.80–88.63)	80.19 (69.99–91.89)	56.62 (47.77–67.11) [a]
≥61	136 (30.8)	0.11 (0.08–0.15) [a]	0.43 (0.30–0.61)	0.81 (0.59–1.12) [a]	1.20 (0.77–1.86) [a]	59.21 (50.80–69.00)	62.58 (53.89–72.69)	66.29 (57.39–76.56)	44.56 (37.23–53.34) [a]
p-value		0.013	0.053	0.004	0.006	0.120	0.116	0.151	0.049
Drinking water									
Ground water/local drinking water	259 (58.6)	0.11 (0.09–0.14)	0.31 (0.23–0.41)	0.63 (0.49–0.81)	0.99 (0.71–1.40)	65.16 (57.90–73.34)	68.68 (61.19–77.09)	71.40 (63.88–79.80)	53.82 (46.84–61.83)
Tap water/purified water	183 (41.4)	0.09 (0.07–0.12)	0.30 (0.22–0.42)	0.52 (0.39–0.70)	0.81 (0.54–1.21)	72.54 (63.03–83.49)	75.89 (66.15–87.06)	78.47 (68.75–89.56)	56.35 (47.78–66.46)
p-value		0.119	0.916	0.207	0.308	0.129	0.149	0.156	0.580
Seafood intake in the last week (missing *n* = 117)									
Yes	268 (82.5)	0.09 (0.07–0.12)	0.26 (0.20–0.33) [a]	0.48 (0.38–0.61) [a]	1.05 (0.71–1.55)	69.31 (61.23–78.45)	72.83 (64.50–82.23)	74.36 (65.97–83.82)	56.86 (50.24–64.35)
No	57 (17.5)	0.14 (0.09–0.23)	0.42 (0.27–0.64) [a]	0.85 (0.57–1.28) [a]	0.79 (0.40–1.55)	64.99 (52.60–80.29)	68.53 (55.70–84.30)	71.04 (57.91–87.15)	52.49 (42.50–64.83)
p-value		0.085	0.022	0.005	0.401	0.542	0.555	0.653	0.448
Distance from mine (km) (missing *n* = 48)									
< 0.5	95 (24.1)	0.12 (0.08–0.17)	0.52 (0.36–0.76) [a]	0.96 (0.68–1.36) [a]	1.31 (0.81–2.12)	52.00 (44.04–61.39) [bc]	55.65 (47.29–65.49) [b]	59.08 (50.50–69.11) [c]	38.95 (32.16–47.16) [b]
0.5–< 1.0	95 (24.1)	0.10 (0.07–0.14)	0.36 (0.24–0.53) [ab]	0.66 (0.46–0.95) [ab]	0.86 (0.52–1.44)	76.36 (64.02–91.07) [b]	79.59 (66.97–94.60) [b]	82.65 (69.98–97.62) [b]	60.51 (49.39–74.12) [ab]
1.0– < 1.5	95 (24.1)	0.09 (0.06–0.14)	0.32 (0.21–0.49) [ab c]	0.56 (0.38–0.83) [ab]	0.78 (0.46–1.32)	76.12 (63.32–91.51) [b]	79.39 (66.28–95.09) [b]	82.62 (69.43–98.31) [b]	62.44 (50.51–77.20) [ab]
1.5– < 3.0	80 (20.3)	0.08 (0.06–0.12)	0.21 (0.14–0.33) [bc]	0.40 (0.27–0.58) [b]	0.73 (0.43–1.24)	81.41 (67.67–97.94) [a]	84.76 (70.71–101.60) [a]	86.06 (72.27–102.48) [a]	71.29 (57.61–88.21) [a]
≥3.0	29 (7.4)	0.09 (0.05–0.16) [c]	0.11 (0.06–0.22) [c]	0.28 (0.15–0.51) [b]	0.60 (0.26–1.38)	49.14 (36.90–65.44) [c]	51.59 (38.96–68.32) [c]	51.86 (39.57–67.98) [d]	46.22 (33.23–64.29) [ab]
p-value		0.587	<0.001	<0.001	0.287	<0.001	<0.001	<0.001	<0.001
As (3+) + As (5+) + MMA + DMA									
1Q (12.60–49.27 µg/g cr)	111 (25.1)	0.08 (0.06–0.11) [a]	0.46 (0.32–0.67) [a]	0.74 (0.53–1.04)	0.74 (0.47–1.17)	26.72 (25.00–28.56) [c]	28.67 (26.88–30.58) [c]	30.96 (29.14–32.90) [c]	22.20 (19.65–25.08) [c]

2Q (49.34–82.17 μg/g cr)	111 (25.1)	0.10 (0.07–0.14)	a	0.30 (0.21–0.45)	ab	0.57 (0.40–0.81)	1.10 (0.68–1.76)	59.90 (55.91–64.19)	b	63.04 (58.96–67.40)	b	64.39 (60.45–68.59)	b	45.98 (40.50–52.19)	b
3Q (80.33–120.28 μg/g cr)	110 (24.9)	0.09 (0.07–0.13)	a	0.23 (0.16–0.34)	b	0.43 (0.31–0.60)	1.06 (0.67–1.67)	92.35 (86.38–98.73)	b	95.90 (89.89–102.32)	b	98.16 (92.34–104.34)	b	74.66 (66.04–84.41)	b
4Q (120.85–633.97 μg/g cr)	110 (24.9)	0.14 (0.10–0.20)	a	0.25 (0.17–0.36)	ab	0.57 (0.40–0.81)	0.79 (0.49–1.26)	170.48 (159.15–182.62)	a	176.29 (164.92–188.44)	a	179.01 (168.10–190.62)	a	134.45 (118.50–152.55)	a
p-value		0.050		0.014		0.075	0.309	<0.001		<0.001		<0.001		<0.001	

abc: Grouping by Bonferroni post-hoc. Estimates with the same letter are not significantly different. Adjusted: least square means adjusted by sex, age, period of residence (missing $n = 1$), drinking water, smoking status, and drinking status.

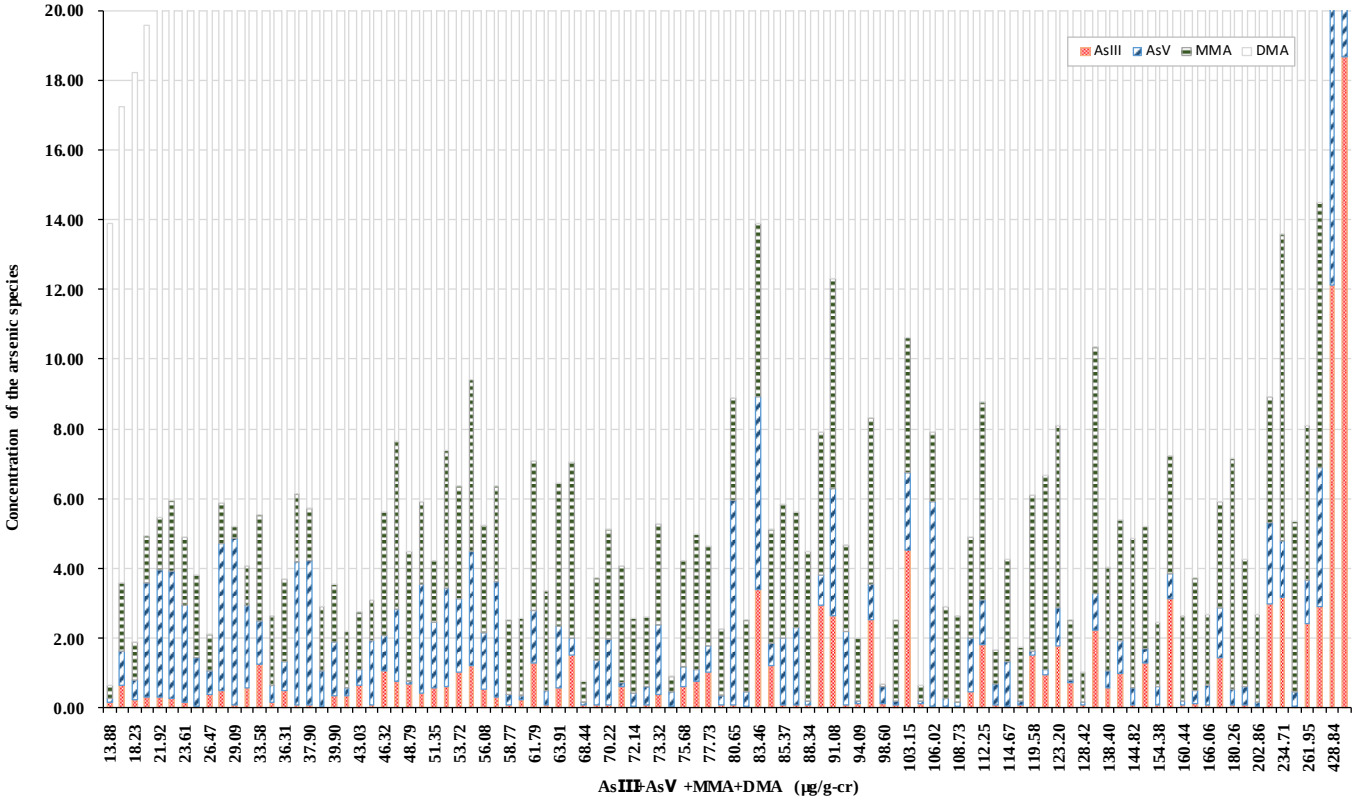

Figure S1. Distribution of As species concentrations by AsIII + AsV + MMA + DMA percentile.

Table S2. Correlation coefficient of As species concentration and proportion.

Group	As Species	HPLC-ICP-MS							HG-AAS	Proportion of As Species Relative to the Sum of Concentrations of 4 As Species						
		Inorganic As			Organic As					Inorganic As			Organic As			
		AsIII	AsV	Sub-total	MMA	DMA	Subtotal	Sum of 4 As Species		AsIII	AsV	Subtotal	MMA	DMA	Subtotal	Sum of 3 As Species

103 mines (n = 457)	HPLC-ICP-MS	Inorganic As	AsIII	1	0.549	0.919	0.822	0.313	0.384	0.479	0.458	**0.763**	0.049	0.350	0.328	-0.410	-0.350	0.410
			AsV		1	0.835	0.681	0.208	0.269	0.360	0.234	0.328	**0.524**	0.599	0.293	-0.579	-0.599	0.579
			Subtotal			1	0.864	0.304	0.381	0.485	0.412	0.658	0.280	**0.514**	0.355	-0.544	-0.514	0.544
		MMA					1	0.358	0.446	0.530	0.436	0.527	0.062	0.267	**0.545**	-0.447	-0.267	0.447
		Organic As	DMA					1	0.995	0.979	0.891	0.079	-0.235	-0.179	-0.188	**0.219**	0.179	-0.219
			Subtotal						1	0.993	0.899	0.130	-0.219	-0.144	-0.124	0.164	**0.144**	-0.164
		Sum of 4 As species								1	0.902	0.205	-0.172	-0.071	-0.073	0.086	0.071	-0.086
	HG-AAS										1	0.207	-0.230	-0.122	-0.120	0.145	0.122	-0.145
	Proportion of As species relative to the sum of concentrations of 4 As species		AsIII									1	0.057	0.453	0.437	-0.536	-0.453	0.536
		Inorganic As	AsV										1	0.916	0.199	-0.772	-0.916	0.772
			Subtotal											1	0.353	-0.905	-1.000	0.905
		MMA													1	-0.718	-0.353	0.718
		Organic As	DMA													1	0.905	-1.000
			Subtotal														1	-0.905
		Sum of 3 AS species																1